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RADIOISOTOPE ACTIVITIES IN URANIUM TAILINGS REFERENCE MATERIALS UTS-1 TO UTS-4: RESULTS OF AN INTERLABORATORY PROGRAM

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RADIOISOTOPE ACTIVITIES IN URANIUM TAILINGS REFERENCE MATERIALS UTS-1 TO UTS-4: RESULTS OF AN INTERLABORATORY PROGRAM

by

C.W. Smith*

ABSTRACT

The results of an interlaboratory measurement program to establish isotope activity values in four compositional uranium-tailings reference materials are described. The materials were prepared from existing Canadian mine-mill tailings from Bancroft Ontario, Elliot Lake Ontario, Beaverlodge Saskatchewan and a pilot-plant tailing of Rabbit Lake Saskatchewan ore (UTS-1 to UTS-4), under the auspices of the National Uranium Tailings Research Program.

Seven laboratories performed multiple determinations of thorium-230, radium-226, lead-210, and polonium-210 in all materials, and four of these determined thorium-232, radium-228 and thorium-228 in UTS-1 and UTS-2. Recommended activity values were determined as the consensus means of individual determinations, and their statistical uncertainties estimated. Results for a control material (UTS-5) suggest that the results are unbiased within their uncertanties. Thorium-230 was found to be depleted relative to radium-226 in UTS-3 and UTS-4; uranium-238 series isotopes activity ratios were otherwise unity within the 95% confidence limits in all materials, as were thorium-232 series isotope ratios.

Analysis of variance indicates that between-laboratories contributions are significant; however, within-laboratories uncertainties generally mask any systematic differences. Average within-laboratory relative standard deviations (RSD) were 10, 6, 7, and 5% respectively for thorium-230, radium-226, lead-210 and polonium-210 whereas RSDs for the recommended values were 17, 10, 13 and 12%. No correlation of precision with activity (3.5 to 35 Bq g^{-1}) was noted except in the case of thorium-230.

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ACTIVITÉS DES RADIO-ISOTOPES DANS DES RÉSIDUS D'URANIUM SERVANT DE MATÉRIAUX DE RÉFÉRENCE UTS-1 À UTS-4: RÉSULTATS D'UN PROGRAMME INTERLABORATOIRES

par

C.W. Smith*

RÉSUMÉ

Les résultats d'un programme interlaboratoires de mesure visant à attribuer des valeurs aux activités isotopiques à l'intérieur de quatre compositions différentes de résidus d'uranium servant de matériaux de référence sont décrits. Les matériaux ont été préparés à partir de résidus provenant de mines et d'usines canadiennes déjà en place à Bancroft et à Elliot Lake en Ontario et à Beaverlodge en Saskatchewan, et à partir de résidus d'une usine pilote de traitement des minerais située à Beaverlodge en Saskatchewan (UTS-1 à UTS-4), sous les auspices du Programme national de recherche sur les résidus d'uranium.

Sept laboratoires ont effectué des mesures multiples pour découvrir du thorium-230, du radium-226, du plomb-210 et du polonium-210 dans tous les matériaux et quatre d'entre eux ont découvert du thorium-232, du radium-228 et du thorium-228 dans l'UTS-1 et l'UTS-2. On a attribué des valeurs aux activités à titre de moyen unique d'effectuer des mesures individuelles, et les incertitudes statistiques de ces valeurs ont été évaluées. Les résultats applicables à un matériau de contrôle (UTS-5) portent à croire que les résultats sont non faussés dans les limites de leurs incertitudes. On a découvert que le thorium-230 était appauvri par rapport au radium-226 dans l'UTS-3 et l'UTS-4 et qu'il y avait unité dans les rapports d'activités isotopiques de la série uranium-238 dans tous les matériaux dans les limites de confiance de 95 %, tout comme dans le cas des rapports isotopiques de la série thorium-232.

L'analyse de variance indique que les contributions entre laboratoires sont importantes; cependant, les incertitudes qui règnent à l'intérieur d'un même laboratoire dissimulent habituellement des différences systématiques. Les écarts-types relatifs moyens à l'intérieur d'un laboratoire étaient de 10, 6, 7 et 5 % respectivement pour le thorium-230, le radium-226, le plomb-210 et le polonium-210, alors que les écarts-types relatifs pour les valeurs recommandées étaient de 17, 10, 13 et 12 %. Aucune corrélation entre la précision et l'activité (de 3,5 à 35 Bq g^{-1}) n'a été notée, sauf dans le cas du thorium-230.

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CONTENTS

	Page
ABSTRACT	i
RÉSUMÉ	ii
INTRODUCTION	1
INTERLABORATORY PROGRAM FOR ISOTOPES	2
CONSENSUS ACTIVITY VALUES: STATISTICAL TREATMENT OF RESULTS	2
DISCUSSION	3
Results for UTS-5	3
Results for UTS-1 to UTS-4	4
Comments on Precision and Accuracy	5
CONCLUSION	9
ACKNOWLEDGEMENTS	10
REFERENCES	11

TABLES

No.		
1.	Contributing laboratories for radiochemical determinations	13
2.	Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-1	15
3.	Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-2	17
4.	Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-3	19
5.	Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-4	21
6.	Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-5	23
7.	Analytical results, laboratory means and standard deviations for ^{232}Th-series isotopes in UTS-1	25
8.	Analytical results, laboratory means and standard deviations for ^{232}Th-series isotopes in UTS-2	26

CONTENTS (Cont'd)

Page

9. Analytical results, laboratory means and standard deviations for ^{232}Th-series isotopes in UTS-5 27
10. Analytical results and laboratory mean values for ^{232}Th and ^{228}Th in UTS-3 and UTS-4 (Lab-3 only, alpha spectroscopy) 28
11. Analytical results and laboratory mean values for ^{231}Pa (Lab-3 only, alpha spectroscopy 28
12. Summary of radiochemical procedures (a,b) 29
13. References cited for radiochemical procedures 31
14. Calibration reference materials and recovery tracers used for radiochemical measurements 33
15. Results of analysis of variance, ^{238}U-series isotopes 34
16. Results of analysis of variance, ^{232}Th-series isotopes 35
17. Results of Dixon r tests applied to laboratory mean results 36
18. Consensus values and related statistical parameters for isotope activities in tailings reference materials 37
19. Radioisotope consensus program performance on UTS-5 39

INTRODUCTION

A project to produce uranium tailings compositional reference materials typical of several existing Canadian tailings sites was initiated under the auspices of the National Uranium Tailings Research Program (NUTRP) in summer of 1982 (1). These are intended to be used in analytical quality-control functions for both chemical and radiochemical measurements within the Measurement Program aspect of the NUTRP. The materials produced will also be made available through the Canadian Certified Reference Materials Project (CCRMP).

This report presents the results of an interlaboratory project to establish the activities of important radioactive isotopes in the reference materials UTS-1 to UTS-4. These results will be incorporated in a comprehensive report including detailed descriptions of the nature, preparation, and results of a separate interlaboratory program to establish the concentrations of selected elements.

UTS-1 is derived from Madawaska Mines mill tailings, Bancroft, Ontario; UTS-2 from Rio Algom mill tailings, Elliot Lake, Ontario; UTS-3 from the now closed Beaverlodge Saskatchewan operations of Eldorado Nuclear Ltd.; and UTS-4 from pilot-plant tailings from Eldor Mines ore, Rabbit Lake, Saskatchewan. The Ontario tailings derive from uranium/thorium mineral-bearing ores while the Saskatchewan ores contain 'trace' levels of thorium.

A fifth material, designated UTS-5, was incorporated in the interlaboratory program. This material was produced by blending the CCRMP compositional reference ore (DH-1a) that contains certified amounts of uranium, thorium, and radium-226 (2,3). Secular equilibrium in both the uranium and thorium decay series is probable. Consequently, UTS-5 was incorporated as a 'yardstick' of the probable quality of the interlaboratory program, and also to test the feasibility of silica dilution for possible future quality control applications. Participants were not informed of the nature of UTS-5.

The radioactive isotopes measured in this project were thorium-230, radium-226, lead-210, and polonium-210 (uranium-238 decay series); and thorium-232, radium-228, and thorium-228 (thorium-232 decay series, measured in UTS-1, UTS-2 and UTS-5 only). In addition, one laboratory provided measurements of thorium-232 and thorium-228 in UTS-3 and UTS-4, and protactinium-231 in all materials.

INTERLABORATORY PROGRAM FOR RADIOISOTOPES

Eight Canadian laboratories contributed isotope activity measurements, seven of these under contract arrangements with Supply and Services Canada (Table 1). All participants were experienced with radiochemical determinations in uranium tailings.

The contract work statement requested that isotope activities (becquerels per gram) be measured in four independently-treated subsamples of each material. Recognized reference standards having published total uncertainties (including a precision estimate at the 99% confidence level) of less than 5% were to be employed in determining instrumental calibration factors for the measurements. Sufficient count data was to be accumulated that the uncertainty component in each result due to counting statistics was less than 2% (at 1σ confidence). Participants were asked to date the determinations of polonium-210 in the event that this isotope proved to be out of secular equilibrium with lead-210. The expertise of the participants was otherwise relied upon to provide unbiased and precise results within their capabilities.

The analytical results are presented in Tables 2-11. The calibration standards are identified in Table 14. Radiochemical methodologies are summarized in Table 12; contractor reports and references cited should be consulted for more detailed descriptions.

CONSENSUS ACTIVITY VALUES: STATISTICAL TREATMENT OF RESULTS

The grand mean of all individual determinations exclusive of statistical outlier sets is defined as the consensus value for the activity of an isotope for purposes of this program. The individual determinations are presumed to be a random sampling of the true value. A statistical model described by Brownlee as 'one-way classification model II' is assumed to valid (4). The application of this model is outlined in various CCRMP publications (2). The uncertainty in the grand mean arising from withinand between-laboratory variances is estimated by the formulae described in these references.

Dixon's 'r' statistic for the extreme laboratory mean values was used to identify laboratory sets having a low probability of belonging to the assumed consensus universe (5). A level of significance $\alpha = 0.05$ ($r_{.05}$) was used for this purpose. The suspected sets were excluded from the calculation

of the consensus value and its uncertainty. Three instances of such apparent outliers at this confidence level were detected in the 29 consensus determinations herein (Table 17). However, the positive result in one case (Ra-228 in UTS-1) was deemed to arise from a possibily fortuitously small variance in the small set of peer means, and was consequently not declared as an outlier set. With one exception (Pb-210 in UTS-3), no other instance of an extreme laboratory mean exceeding the 10% critical value was observed. In this case, the set was deleted for purposes of calculation of the consensus mean, since it would otherwise inordinately weight the value due to the relatively large number of individual determinations in this set. The set was retained, however, for purposes of estimation of the uncertainty in the consensus value in this case.

Results of analysis of variance (Tables 15 and 16) indicate that the between-laboratory component is generally significant relative to the within-laboratory component as has been observed to be the norm in chemical analysis consensus programs.

The consensus activity values for UTS-1 to UTS-4, their uncertainty estimates, and associated statistical parameters are compiled in Table 18. The confidence limits (CL) estimate the range of reproducibility of the mean in 95% of cases were the identical program to be repeated many times. The average within-laboratory relative standard deviation ($\overline{CV}$, %) and the relative standard deviation of and individual determination from the grand mean (RSD, %) provide an indication of how closely an individual determination approaches respectively the laboratory estimate and consensus value on the average for general comparative purposes.

The corresponding results for UTS-5 are compiled separately in Table 19; a 'bias estimate' comparing the consensus and predicted activity values is included for this material.

DISCUSSION

RESULTS FOR UTS-5

. UTS-5 was incorporated in the consensus program as a 'blind control' material to indicate potential biases in laboratory methods and consequently in the consensus values. Gamma-ray and uranium concentration measurements indicated that a homogeneous dilution of DH-1a (49.4 ± .6%, 2σ) in the silica

was accomplished in the blending of this material (6). From the certified uranium and thorium concentrations in DH-1a and published half-lives, the activities of uranium-238, uranium-235 and thorium-232 in UTS-5 are 15.86 ± .32, 0.744 ± .018, and 1.82 ± .10 Bq g^{-1} respectively, with uncertaintly estimates propagated at the 99% confidence level (2,7-9). The consensus value for radium-226 in DH-1a in a thirteen-laboratory consensus program was 31.5 ± 1.1 Bq g^{-1} essentially confirming uranium-radium secular equilibrium (3). It is inferred that there is a high probability that thorium-230 is also in secular equilibrium. Radon emanation rate measurements of DH-1a suggest that lead-210 and polonium-210 may be 3 to 5% or more below secular equilibrium values if these rates existed in the natural ore state (10-12). Polonium-210 in DH-1a was measured as 31.3 ± 1.1 Bq g^{-1} (95% CL), however, suggesting equilibrium (12). In view of the relatively short half-lives of radium-228 and thorium-228 and the nature of DH-1a, it is highly probable that these isotopes are in secular equilibrium with thorium-232.

The consensus program results for UTS-5 are compared with the predicted (secular equilibrium) conditions in Table 19. In all cases, the consensus means are in agreement with the predicted values, within the 95% confidence limits. The 'bias estimate', i.e., the relative departure of the consensus mean from the predicted value, is well within the 95% confidence interval estimate for each isotope. The bias estimate approaches the 95% confidence limit only for polonium-210. The single laboratory result for protactinium-231 suggests either a slightly low methodological bias or a lower than equilibrium activity for this isotope in UTS-5.

It is inferred that the consensus means, within the statistically determined confidence intervals, are likely to provide reliable estimates of the activities present in the tailing materials, provided that very different matrix or spectral interference effects are not encountered.

RESULTS FOR UTS-1 TO UTS-4

The recommended values for isotope activities for the tailings reference materials UTS-1 to UTS-4 are compiled in Table 18.

The recommended activities of uranium-238 decay series isotopes in both UTS-1 and UTS-2 are not significantly different, i.e., their 95% confidence intervals overlap at about 3.5 and 5.1 Bq g^{-1} respectively. Thus thorium-230, radium-226, lead-210, and polonium-210 are close to, if not in, secular equilibrium in UTS-1 and UTS-2.

Thorium-230 is found to be depleted relative to radium-226 in UTS-3 and UTS-4 with activity ratios of .85 ± .07 and .59 ± .08 (2σ) respectively. Thorium-230 is similarly depleted relative to lead-210 in UTS-4 (Th/Pb .66 ± .11). Within the measurement uncertainties, polonium-210 is supported by lead-210 in both materials (activity ratios of .94 ± .23 and .89 ± .18 respectively). Similarly lead-210 may be completely supported by radium-226 (activity ratios of .95 ± .25 and .90 ± .12 respectively).

There is no significant evidence for disequilibrium between thorium-232, radium-228, and thorium-228 in UTS-1 or UTS-2. It was decided to incorporate only the α-spectrometry measurements of thorium-232 in the radiochemistry consensus program, as neutron activation measurements of thorium are being incorporated in the chemical analysis certification program for these materials. Thorium-232 activities herein should be compared with those calculable from the determined concentrations.

COMMENTS ON PRECISION AND ACCURACY

The confidence intervals generally indicate the current extent of agreement between participating laboratories as to the activities that are present in these Canadian tailings materials. On the average (over the five materials) the relative 95% confidence intervals are radium-226 (±6.5%), thorium-230, lead-210, and polonium-210 (all about ±13%), thorium-232 by alpha spectrometry (±19%), thorium-228 (±24%), and radium-228 (±34%)*.

The relative standard deviations of individual values from the consensus means provide a comparative measure of how closely a single determination approaches an interlaboratory consensus value at present (Tables 18 and 19). Averaged over the five materials, these are 17, 10, 13 and 12% respectively for thorium-230, radium-226, lead-210, and polonium-210. Averaged over the three materials the values are 19, 20 and 22% for thorium-232 (alpha-spectrometry), radium-228, and thorium-228.

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*The confidence intervals are of course weighted by the numbers of determinations and laboratories which are considerably different for the uranium-238 series and thorium-232 series isotopes in this program (4,5).

Similarly, the values of average within-laboratory relative standard deviation ($\overline{CV}$, %) indicate how closely an individual determination may be expected to approach the mean value of a number of measurements within an 'average' participating laboratory. Averaged over the materials, the values are 10, 6, 7 and 5% respectively and 12, 11 and 10% respectively for the uranium-238 series and thorium-232 series as above.

With the exception of lead-210 determinations, no consistent evidence for relative methodological bias between laboratories could be deduced from an examination of laboratory mean values for all materials. In the case of lead-210 the mean results from laboratories 2 and 5 were consistently the highest in all materials. If relative bias exists in other determinations, it is sufficiently small to be masked by the within-laboratory mean uncertainties. Some partial trends are observed, however, these are inconclusive in view of the small amount of data. That the laboratory mean values are otherwise apparently randomly distributed lobbies in favour of the hypothesis that the consensus mean, within its statistical uncertainty, is the best available estimator of the true value from the program data. The validity of this approach is supported by the results for UTS-5.

Note must be taken, however, of the results found to lie outside of the consensus, particularly in view of the strong between-laboratory components of variance and the statistically small number of laboratories involved in the program. Inclusion of sets declared as outliers would result in recommended values remaining within the current 95% confidence intervals, but would approximately double these uncertainties. The apparent disagreements do not appear to stem from consistently observed methodological or calibration differences.

The relatively high degree of between-laboratory accord in the determinations of radium-226 is noteworthy. This probably arises from a combination of the use of established separation and tracer methodologies and a generally higher degree of experience with the isotope than with others, the use of common-source high-precision calibration standards, and possibly a less idiosyncratic nature of radium-226 towards matrix and spectral interference problems.

The accuracy of the results herein of course can be no better than the accuracy of the instrumental calibrations by which they were obtained. Calibrations, for the most part, were obtained using commercially available

isotope reference materials bearing certified total uncertainties (Table 14). The uncertainties in the calibration standards are not explicitly incorporated in the statistical uncertainties reported herein. Any significant differences in calibration parameters, laboratory-to-laboratory, would of course be reflected in the between-laboratories variance and hence be incorporated in the recommended value uncertaintly. As has already been mentioned, such consistent biases are apparently masked by larger within-laboratory variances with one exception.

The NBS radium standards used in the present work are certified as to radium mass, however, the uncertaintly estimated for the specific activity and half-life of radium-226 of ±0.44% (3σ) increases the relative uncertainty in the calculated activity only slightly (13).

Calibration solutions containing thorium-232, radium-228 and thorium-228 prepared from the 1906-refined thorium nitrate hydrated salt bear activity uncertainties propagated from uncertainties in the mass of thorium present* and the specific activity of thorium-232 (8,9). The activity uncertainties of all three isotopes are probably less than 5% in view of the age of the salt. This material (AM TYS-1) is employed as a calibration source for the alpha-spectrometry determinations of thorium isotopes and the beta-counting determination of radium-228 by several participants.

Laboratory 4 determined the activity of a thorium-228 solution relative to that of thorium-232 in a thorium nitrate solution from alpha activity ratios measured in a standard additions procedure. The thorium-230/thorium-228 activity ratios in spiked samples were then used to deduce thorium-230 activities. The ratios in unspiked samples were used to correct for the natural thorium-228 present, however, this interference was negligible except in the case of UTS-5. The activities of thorium-232 and thorium-238 in UTS-1, UTS-2 and UTS-5 were determined relative to thorium-230 from the alpha activity ratios from the unspiked samples.

Secular equilibrium assumptions are implicit in the use of chemical calibration standards by laboratory 1 (monazite sand for thorium-228 and radium-228), laboratory 4 (aged thorium oxide for radium-228), and laboratory

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*One lot of this material was found to contain about 3.4% of sulphate and 40.75 ± .05% thorium vs 39.45% for the formulation $Th(NO_3)_4\ 6H_2O$ (14).

7 (BL-5 for thorium-230). The assumptions in the former two cases are probably reasonable from age considerations, and in the latter case by inference from indicated radium-226/uranium-238 secular equilibrium (3).

Laboratory 8 employed commercial americium-241 and lead-210 thin sources to determine the geometry/efficiency calibration for its alpha spectrometer and beta proportional counter respectively. Calibrations were confirmed with a standard radium-226 solution and other sources. This laboratory reported a problem with apparent changes in the activity concentration of radium-226 standard solution stored in a polyethelene bottle.

Calibration standards used for the determinations of lead-210 and polonium-210 were otherwise exclusively performed with commercial certified solutions of lead-210/bismuth-210/polonium-210 (AM RBZ), polonium-210, or polonium-208. Polonium is notorious for plating out on containers and the RBZ reference solution certificate bears a caveat that there is no guarantee that the polonium remains in solution. If this occurred in calibrating solutions but not correspondingly in samples, a high bias in reported values would result. This is not indicated in the results for UTS-5, however, laboratory 5 observed glassware contamination by polonium during some determinations, which was erratically released in subsequent useage.

No obvious general correlation was noted between the magnitude of activity (or nature of the material) and the magnitude of uncertainty of a given isotope. Laboratory 5 indicated that within-laboratory reproducibilities experienced for thorium-230 and radium-226 in UTS-4 were poorer than expected, and that gamma-ray measurements of radium-226 in UTS-3 and UTS-4 differed significantly from alpha spectrometry results, both for unknown reasons. Similarily, laboratory 6 reported 'poorer-than-expected' precisions for extended replicate determinations of radium-226 and lead-210 in UTS-2. Laboratory 8 found that natural barium in some materials degraded their alpha spectral resolution sufficiently that tailing corrections were required, raising uncertainties somewhat.

CONCLUSION

The activities of four uranium-238 decay series isotopes in the Canadian tailings reference materials UTS-1 to UTS-4 and three thorium-232 series isotopes in UTS-1 and UTS-2 are established herein by consensus of participants who represent a considerable body of experience in such radiochemical determinations. The agreement between measured and predicted values for the control material UTS-5 suggests that the consensus results are likely to be unbiased, and that the statistical confidence intervals reliably estimate the uncertainties.

There is a dearth of available data from interlaboratory isotope measurement programs. Radium-226 has been measured in ore materials by the CCRMP, and a similar project for lead-210 has been initiated* (3). Similarly the U.S. Department of Energy conducted an interlaboratory radium-226 measurement project for ore materials (15). The Canadian Uranium Producers Metallurgical Committee, Analytical Subcommittee carried out an unpublished interlaboratory study of radium-226 in CCRMP ores (16). The U.S. Environmental Protection Agency has carried out an extensive laboratory certification program with isotope containing solutions, and has developed tailings standards, but the availability of interlaboratory data is unknown to this author (17). The relative uncertainites for radium-226 found in the present tailings program are in line with those observed for ore materials in the CCRMP project which was more extensive in terms of numbers of participants.

The statistical information presented herein indicates the probable magnitudes of uncertaintly present in a given isotopic activity measurement by presently accepted methodology. Knowledge of these uncertainties is important both for the statistical component of design of a tailings research measurement program, and as a potential limitation on conclusions which may be drawn from such a program.

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Table 1 - Contributing laboratories for radiochemical determinations*

Lab-1**	Nuclear Activation Services Hamilton, Ontario (E.L. Hoffman)
Lab-2**	Chemex Laboratories Ltd. North Vancouver, British Colombia (M. Legeyt)
Lab-3	Monenco Analytical Laboratories Calgary, Alberta (J. Dean, N. Chiu) Contract 15SQ.23440-2-9145-4
Lab-4	Physics Department, University of Calgary Calgary, Alberta (C.J. Bland, P. Jarvis) Contract 15SQ.23440-2-9145-5
Lab-5	Saskatchewan Research Council Saskatoon, Saskatchewan (G. Smithson, V. Penner, M. Knelson, R. Ortlepp, L. MacDonald) Contract 15SQ.23440-2-9145-1
Lab-6	Eldorado Resources Ltd., Research and Development Division Ottawa, Ontario (M.C. Miedema, G.A. Dunlop, R. Jones, G.B. Moodie) Contract 15SQ.23440-2-9145-2
Lab-7	CANMET, Mineral Sciences Laboratories, Chemical Laboratory Ottawa, Ontario (J.L. Dalton, R. McCorkell, M. DeGagne)

Table 1 (Cont'd)

Lab-8	Waterloo Research Institute, University of Waterloo Waterloo, Ontario (H. Sharma, B. Hauk) Contract 15SQ.23440-2-9145-6

*Supply and Services Canada contracts as indicated, contractor reports filed with CANMET Research Program Office and National Uranium Tailings Research Program Office.

**Sub-contracted by Materials Research Laboratories Ltd., Nepean, Ontario (S.K. Singh), contract 15SQ.23440-2-9145-3.

Table 2 - Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-1

		Bq g^{-1}						Mean	S.D.
^{230}Th	Lab-2 (α)	2.37	2.74	2.59	2.48			2.545	.158
	Lab-3 (α)	3.52	4.81	3.52	5.37			4.305	.935
	Lab-4 (α)	4.91	4.56	3.70	4.37			4.385	.509
	Lab-5 (α)	3.83	3.46	4.04	3.64			3.743	.249
	Lab-6 (α)	3.01	3.15	3.21	3.02			3.098	.098
	Lab-7 (α)	2.59	3.77	4.11	3.29			3.440	.659
	Lab-8 (α)	3.50	3.63	3.77	3.79			3.673	.135
^{226}Ra	Lab-1 (α)	2.9	3.3	4.8	3.7			3.68	.82
	Lab-3 (α)	4.03	3.81	3.42	3.59			3.713	.265
	Lab-4 (α)	3.88	3.60	3.80	4.13			3.853	.219
	Lab-5 (gα)	3.53	3.53	3.47	3.48	3.49		3.500	.028
	Lab-5 (γ)	3.49	3.52					3.505	.021
	Lab-6 (Rn)	3.38	3.65	3.44	3.33			3.450	.141
	Lab-7 (α)	3.81	3.40	3.96	3.85			3.755	.245
	Lab-8 (α)	3.79	4.04	3.59	4.15			3.893	.252
^{210}Pb	Lab-2 (β)	3.52	3.44	3.48	3.52			3.490	.038
	Lab-3 (β)	3.00	3.03	3.18	3.00			3.052	.086
	Lab-4 (α, ^{210}Po)	3.33						3.33	
	Lab-5 (β)	3.33	3.33	3.51	3.63	3.66	3.66	3.520	.157
	Lab-6 (β)	3.27	3.24	3.23	3.15			3.223	.051
	Lab-7*(β)	2.52	2.40	2.52	1.81			2.313	.340
	Lab-8 (β)	3.70	2.72	4.08	3.34			3.460	.579

Table 2 (Cont'd)

		Bq g^{-1}						Mean	S.D.
^{210}Po	Lab-2 (a)	3.44	3.22	3.26	3.03			3.238	.168
	Lab-3 (a)	2.8	2.9	2.8	2.8			2.82	.05
	Lab-4 (a)	3.60	3.77	3.46	3.34			3.542	.185
	Lab-5 (a)	2.92	2.92	2.89	2.81	2.89	2.96		
		2.81						2.886	.057
	Lab-6 (a)	3.00	2.81	3.03	2.85			2.922	.109
	Lab-8 (a)	3.48	3.44	3.67	3.66			3.562	.120

*outlier set

Table 3 - Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-2

		Bq g^{-1}						Mean	S.D.
^{230}Th	Lab-2 (α)	3.26	3.22	3.44	3.33			3.312	.096
	Lab-3 (α)	5.84	3.57	5.96	4.70			5.108	1.120
	Lab-4 (α)	4.96	4.06	4.80	4.62			4.610	.392
	Lab-5 (α)	3.87	4.08	3.78	3.80	3.72		3.850	.139
	Lab-6 (α)	3.06	3.33	3.18	3.14			3.179	.113
	Lab-7 (α)	4.74	4.66	4.37	4.63			4.600	.160
	Lab-8 (α)	6.22	6.66	6.77	6.70			6.588	.249
^{226}Ra	Lab-1 (α)	5.9	5.5	5.9	7.3			6.15	.79
	Lab-3 (α)	6.36	4.91	4.74	4.55			5.140	.827
	Lab-4 (α)	6.25	5.84	6.06	6.07			6.055	.168
	Lab-5 (gα)	5.80	5.80	5.89	5.65			5.785	.100
	Lab-5 (γ)	5.64	5.00					5.320	.453
	Lab-6 (Rn)	5.01	6.06	5.06	7.01	4.77	6.19		
		5.68	3.48	5.76	5.13	5.42		5.416	.910
	Lab-7 (α)	6.10	5.66	6.07	6.03			5.965	.205
	Lab-8 (α)	5.35	4.72	5.06	4.88			5.003	.270
^{210}Pb	Lab-2 (β)	4.85	4.70	4.60	5.33			4.870	.323
	Lab-3 (β)	4.40	4.44	4.22	4.29			4.338	.101
	Lab-4 (α, ^{210}Po)	4.51						4.51	
	Lab-5 (β)	5.07	5.55	5.66	5.11	5.44	5.40		
		5.55	5.36	5.55	5.48			5.417	.193
	Lab-6 (β)	5.48	4.19	3.69	3.99	5.17	4.00		
		5.59	4.84	4.66	3.15	3.97		4.430	.778
	Lab-7 (β)	3.77	3.26	2.81	3.96			3.450	.519
	Lab-8 (β)	3.82	3.75	4.75	5.24			4.390	.727

Table 3 (Cont'd)

		Bq g^{-1}						Mean	S.D.
^{210}Po	Lab-2 (a)	4.92	4.81	5.18	4.14			4.762	.443
	Lab-3 (a)	4.2	4.3	3.66	4.5			4.165	.359
	Lab-4 (a)	5.44	5.43	5.69	5.52			5.520	.120
	Lab-5 (a)	3.88	4.25	3.88	3.88	4.37	4.07		
		4.44	4.37	4.00				4.127	.224
	Lab-6 (a)	4.26	4.33	4.66	4.36			4.403	.177
	Lab-8 (a)	4.11	3.85	3.48	3.50			3.735	.302

Table 4 - Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-3

		Bq g^{-1}						Mean	S.D.
^{230}Th	Lab-2 (α)	10.2	10.0	10.5	11.0			10.42	.435
	Lab-3 (α)	11.4	12.1	11.9	12.2			11.90	.356
	Lab-4 (α)	11.6	13.0	13.7	13.2			12.88	.900
	Lab-5 (α)	11.5	11.7	12.6	11.3	11.1		11.64	.581
	Lab-6 (α)	10.9	10.6	10.4	10.4			10.58	.236
	Lab-7 (α)	12.5	10.7	11.1	9.7			11.00	1.160
	Lab-8 (α)	11.0	11.3	10.2	10.4			10.73	.512
^{226}Ra	Lab-1 (α)	14.6	14.6	14.6	7.3			12.8	3.7
	Lab-3 (α)	13.0	14.0	13.4	14.3			13.68	.585
	Lab-4 (α)	14.4	14.9	14.5	13.7			14.38	.499
	Lab-5 (gα)	12.9	12.5	12.8	12.9			12.78	.189
	Lab-5 (γ)	14.6	14.4					14.5	.14
	Lab-6 (Rn)	13.1	12.0	12.8	12.9			12.70	.483
	Lab-7 (α)	13.9	13.2	13.8	13.6			13.63	.310
	Lab-8 (α)	12.2	12.0	12.9	13.4			12.60	.668
^{210}Pb	Lab-2 (β)	13.1	13.9	14.1	13.3			13.60	.476
	Lab-3 (β)	11.5	12.1	12.3	13.2			12.28	.704
	Lab-4 (α, ^{210}Po)	11.4						11.4	
	Lab-5*(β)	15.4	15.9	15.9	16.3	16.3	15.9		
		16.6	15.4	16.4				16.01	.426
	Lab-6 (β)	12.0	12.7	11.9	12.7			12.32	.435
	Lab-7 (β)	10.8	12.8	14.8	9.29			11.92	2.397
	Lab-8 (β)	13.9	13.2	13.1	11.9			13.02	.830

Table 4 (Cont'd)

		Bq g^{-1}						Mean	S.D.
^{210}Po	Lab-2 (α)	13.2	13.6	13.7	12.7			13.30	.455
	Lab-3 (α)	11.7	10.1	11.8	10.9			11.12	.793
	Lab-4 (α)	11.8	11.0	11.2	11.2			11.30	.346
	Lab-5 (α)	11.84	11.84	11.84	11.65	12.02	11.84		
		12.21	12.21	11.47				11.88	.241
	Lab-6 (α)	11.11	9.99	11.03	10.99			10.78	.529
	Lab-8 (α)	12.8	13.1	12.3	12.3			12.62	.395

*outlier set

Table 5 - Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-4

		Bq g^{-1}						Mean	S.D.
^{230}Th	Lab-2 (α)	22.5	23.4	22.8	23.2			22.98	.404
	Lab-3 (α)*	40.8	42.7	36.6	41.5			40.40	2.652
	Lab-4 (α)	24.2	24.2	24.5	23.1			24.00	.616
	Lab-5 (α)	16.6	16.5	18.6	20.6			18.08	1.941
	Lab-6 (α)	24.4	24.6	22.6	23.0			23.65	.998
	Lab-7 (α)	27.2	17.9	27.1	24.6			24.20	4.369
	Lab-8 (α)	23.6	26.2	24.1	24.7			24.65	1.127
^{226}Ra	Lab-1 (α)	36.6	47.6	47.6	40.2			43.0	5.51
	Lab-3 (α)	37.1	40.0	38.7	38.9			38.68	1.196
	Lab-4 (α)	39.4	39.9	40.4	39.1			39.70	.572
	Lab-5 (gα)	39.2	31.2	38.0	31.4	37.1	37.0		
		32.1						35.14	3.433
	Lab-5 (γ)	35.9	35.6					35.75	.212
	Lab-6 (Rn)	39.4	36.5	35.0	39.5			37.60	2.223
	Lab-7 (α)	38.0	39.7	39.3	39.6			39.15	.785
	Lab-8 (α)	40.5	39.4	42.7	40.7			40.82	1.374
^{210}Pb	Lab-2 (β)	36.7	35.3	36.6	37.0			36.40	.753
	Lab-3 (β)	33.0	30.7	32.0	33.7			32.35	1.303
	Lab-4 (α, ^{210}Po)	30.6						30.6	
	Lab-5 (β)	38.8	39.8	39.6	38.5	38.8	42.6		
		41.8						39.99	1.598
	Lab-6 (β)	35.9	35.2	35.9	35.6			35.65	.332
	Lab-7 (β)	30.0	31.4	35.8	32.6			32.45	2.473
	Lab-8 (β)	28.8	28.2	26.9	29.7			28.40	1.175

Table 5 (Cont'd)

		Bq g^{-1}						Mean	S.D.
^{210}Po	Lab-2 (α)	34.8	34.6	34.5	38.0			35.48	1.688
	Lab-3 (α)	23.5	25.4	23.0	24.5			24.10	1.068
	Lab-4 (α)	35.3	37.8	32.1	37.4			35.65	2.608
	Lab-5 (α)	29.6	30.3	28.9	28.1	31.1	27.8		
		29.6	29.6					29.38	1.090
	Lab-6 (α)	33.6	34.0	33.9	36.4			34.48	1.295
	Lab-8 (α)	27.9	27.4	26.9	27.6			27.45	.420

*outlier set

Table 6 - Analytical results, laboratory means and standard deviations for ^{238}U-series isotopes in UTS-5

		Bq g^{-1}						Mean	S.D.
^{230}Th	Lab-2 (α)	15.9	16.5	16.4	16.4			16.30	.271
	Lab-3 (α)	14.6	14.8	14.8	14.1			14.52	.613
	Lab-4 (α)	15.6	15.3	16.0	16.3			15.80	.440
	Lab-5 (α)	15.7	15.6	15.9	15.8	15.3	15.8	15.60	.294
		15.1							
	Lab-6 (α)	15.3	15.7	14.3	15.8			15.28	.685
	Lab-7 (α)	16.1	18.5	16.6	18.2			17.35	1.179
	Lab-8 (α)	13.0	12.4	13.4	12.2			12.75	.551
^{226}Ra	Lab-1 (α)	21.6	22.0	18.3	18.3			20.05	2.027
	Lab-3 (α)	13.9	14.2	14.7	15.3			14.52	.613
	Lab-4 (α)	16.7	16.9	16.5	16.6			16.68	.171
	Lab-5 (gα)	15.3	16.3	16.7	16.6	15.1	15.0	15.83	.784
	Lab-5 (γ)	17.8	17.5					17.65	.212
	Lab-6 (Rn)	16.2	14.8	16.1	15.0			15.52	.727
	Lab-7 (α)	15.6	16.3	15.8	16.5			16.05	.420
	Lab-8 (α)	16.1	15.3	15.1	15.7			15.56	.444
^{210}Pb	Lab-2 (β)	16.4	17.0	17.3	16.9			16.90	.374
	Lab-3 (β)	13.4	13.9	13.8	14.8			13.98	.591
	Lab-4 (α, ^{210}Po)	13.8						13.8	
	Lab-5 (β)	16.8	16.6	16.7	17.0	17.2	17.4		
		17.7						17.06	.399
	Lab-6 (β)	14.3	14.8	14.7	14.7			14.62	.222
	Lab-7 (β)	17.7	13.7	13.0	16.8			15.30	2.299
	Lab-8 (β)	11.4	10.5	9.2	11.9			10.75	1.185

Table 6 (Cont'd)

		Bq g^{-1}						Mean	S.D.
^{210}Po	Lab-2 (α)	15.7	17.7	16.6	17.7			16.92	.967
	Lab-3 (α)	11.3	10.4	11.5	14.2			11.85	1.638
	Lab-4 (α)	14.2	14.8	15.2	14.5			14.68	.427
	Lab-5 (α)	11.5	12.6	13.0	12.6	11.8	11.8		
		13.0						12.33	.618
	Lab-6 (α)	14.5	15.1	16.2	15.5			15.32	.714
	Lab-8 (α)	11.9	12.4	12.6	10.1			11.75	1.139

Table 7 - Analytical results, laboratory means and standard deviations for ^{232}Th-series isotopes in UTS-1

		Bq g^{-1}					Mean	S.D.
^{232}Th	Lab-1 (NAA)	.514	.579	.551	.555		.550	.027
	Lab-2 (α)	.510	.586				.548	.054
	Lab-2 (γ, daught)	.570	.570				.570	
	Lab-3 (α)	.58	.82	.61	.93		.735	.168
	Lab-4 (α)	.810	.774	.614	.717		.729	.086
	Lab-5 (α)	.66	.55	.65	.65		.628	.052
	Lab-5 (NAA)	.61					.61	
	Lab-8 (α)	.75	.70	.73	.68		.715	.031
^{228}Ra	Lab-1 (β)	<1.	<1.	<1.	<1.			
	Lab-3 (β)	.74	.92	.74	.63		.758	.120
	Lab-4 (γ)	.736					.736	
	Lab-5 (β)	.54	.53	.55	.52	.57	.542	.019
	Lab-8 (β)	.88	.71	.69	.75		.758	.085
^{228}Th	Lab-2 (α)	.6	.6	.6	.6		.6	
	Lab-3 (α)	.64	.67	.67	.92		.725	.131
	Lab-4 (α)	.85	.79	.62	.75		.752	.097
	Lab-5 (α)	.65	.58	.65	.65		.632	.035
	Lab-8 (α)	1.22	.78	.77	.66		.858	.248

Table 8 - Analytical results, laboratory means and standard deviations for ^{232}Th-series isotopes in UTS-2

		Bq g^{-1}						Mean	S.D.
^{232}Th	Lab-1 (NAA)	.693	.656	.680	.680			.677	.015
	Lab-2 (α)	.76	.69					.725	.050
	Lab-2 (γ, daught)	.688	.692					.690	.003
	Lab-3 (α)	.57	.65	1.20	.83			.812	.280
	Lab-4 (α)	1.10	.85	.98	.98			.978	.102
	Lab-5 (α)	.56	.68	.85	.84	.77		.740	.121
	Lab-5 (NAA)	.74						.74	
	Lab-8 (α)	.91	.91	1.27	1.26			1.088	.205
^{228}Ra	Lab-1 (β)	<1.	<1.	<1.	<1.			<1.	
	Lab-3 (β)	1.07	1.11	1.26	1.15			1.148	.082
	Lab-4 (γ)	1.26						1.26	
	Lab-5 (β)	.91	.80	.89	.80	.80	.85	.842	.050
	Lab-8 (β)	1.18	1.16	1.23	1.09			1.165	.058
^{228}Th	Lab-1 (α)	.6	.6	.6	.6			.6	
	Lab-3 (α)	.85	.81	1.27	1.18			1.028	.232
	Lab-4 (α)	1.07	.86	1.03	1.00			.990	.091
	Lab-5 (α)	.56	.68	.87	.79	.74		.728	.117
	Lab-8 (α)	1.23	1.29	1.21	1.44			1.293	.104

Table 9 - Analytical results, laboratory means and standard deviations for ^{232}Th-series isotopes in UTS-5

		Bq g^{-1}						Mean	S.D.
^{232}Th	Lab-1 (NAA)	1.89	1.89	1.88	1.86			1.880	.014
	Lab-2 (α)	1.97	1.90					1.935	.050
	Lab-2 (γ, daught)	1.94	1.88					1.910	.042
	Lab-3 (α)	2.07	2.40	1.92	1.85			2.060	.244
	Lab-4 (α)	2.42	2.31	2.34	2.43			2.375	.059
	Lab-5 (α)	1.69	1.72	1.93	1.71			1.762	.112
	Lab-5 (NAA)	1.76						1.76	
	Lab-8 (α)	1.82	1.46	2.01	1.35			1.660	.308
^{228}Ra	Lab-1 (β)	1.0	2.0	1.0	1.0			1.25	.500
	Lab-3 (β)	2.2	2.2	2.1	2.6			2.28	.222
	Lab-4 (γ)	2.95						2.95	
	Lab-5 (β)	1.70	1.59	1.66	1.59	1.66	1.88		
		1.59						1.667	.104
	Lab-8 (β)	1.95	1.73	1.80	1.85			1.833	.092
^{228}Th	Lab-1 (α)	1.8	1.8	1.8	1.8			1.80	
	Lab-3 (α)	1.97	2.40	2.04	1.85			2.065	.237
	Lab-4 (α)	2.55	2.40	2.47	2.55			2.492	.072
	Lab-5 (α)	1.69	1.72	2.02	1.98			1.852	.171
	Lab-8 (α)	1.61	1.53	1.84	1.87			1.712	.168

Table 10 - Analytical results and laboratory mean values for ^{232}Th and ^{228}Th in UTS-3 and UTS-4 (Lab-3 only, alpha spectroscopy)

		Bq g^{-1}				Mean	S.D.
UTS-3	^{232}Th	.15	.16	.22	.10	.16	.05
	^{228}Th	.13	.16	.23	.13	.16	.04
UTS-4	^{232}Th	.40	.56	.73	.48	.48	.07
	^{228}Th	.22	.27	.19	.26	.23	.04

Table 11 - Analytical results and laboratory mean values for ^{231}Pa (Lab-3 only, alpha spectroscopy)

	Bq g^{-1}				Mean	S.D.
UTS-1	.23	.23	.20	.18	.21	.03
UTS-2	.30	.37	.51	.31	.37	.11
UTS-3	.54	.78	.61	.85	.70	.15
UTS-4	2.2	2.3	2.1	2.8	2.4	.2

Table 12 - Summary of radiochemical procedures (a,b)

Lab-1 Decompositions by repeated multiple acid digestion; HCl, HNO_3, HF, H_2SO_4. Residues fused with $LiBO_2$. Dissolved in 10% HNO_3/10% HCl.

Separations: (volumetric aliquots)

^{226}Ra - (c), α-spectroscopy.

^{228}Ra - (c) (no ^{133}Ba), β-counting.

^{232}Th, ^{228}Th - two-stage IX; SX (TTA), extract concentrated and evaporated on a steel planchet; α-spectroscopy; ^{232}Th also by NAA.

Lab-2 Decomposition by repeated multiple acid digestions; HNO_3, HF, $HClO_4$ (0.25 to 0.5 g).

Separations:

^{210}Pb - (d) (1).

^{210}Po - Ag deposition with ascorbic acid reductant (1), α-spectrometry.

^{230}Th - Successive coprecipitations with lanthanum as hydroxide and fluoride, with Ba 'hold-back' carrier for Ra, final ppt. collected on 0.45 μm filter for α-spectrometry. ^{234}Th tracer by β-counting. ^{230}Th additions implied negligible self-absorption.

Lab-3 Decompositions: for Ra, Th: 2 g by KF and pyrosulphate fusion (2).
for Po, Pb: 1 g by repeated multiple digestions; HNO_3, HF, $HClO_4$.

Separations:

^{226}Ra - (c), α-spectrometry.

^{228}Ra - (e) (1,3,4,5), ^{228}Ac β-counting.

^{230}Th, ^{232}Th, ^{228}Th - supernate from (c) volumetrically diluted, and split into two equal portions, one spiked with ^{228}Th. Following SX (HDEHP) (5), thorium coprecipitated with cerous hydroxide, collected on 0.1 μm filter for α-spectrometry (6).

^{210}Pb - (d) (1).

^{210}Po - coprecipitation with PbS, dissolved in HCl, Ni deposition for α-spectrometry (7). ^{208}Po tracer used.

Lab-4 Decompositions by HF treatment followed by KF and pyrosulphate fusions of 1 g samples (8,9).

Separations:

^{226}Ra - (c), α-spectrometry.

^{232}Th, ^{228}Th, ^{230}Th - coprecipitated with cerous hydroxide in presence of EDTA from supernate from (C) filtered onto 0.1 μm filter for α-spectrometry.

^{210}Po - coprecipitated with tellerium in presence SO_2, $TiCl_3$, collected on 0.1 μm filter. (^{208}Po tracer used, α-spectrometry.)

^{228}Ra - no separations, γ-spectrometry of ^{228}Ac on 40-75 g samples.

^{210}Pb - indirectly, as supported ^{210}Po.

Lab-5 Decompositions of 1 g samples by repeated digestion with HF, H_2SO_4 followed by pyrosulphate fusions. Radium samples dissolved in HNO_3, others in HCl.

Separations:

^{226}Ra - (c) (1) (gross-alpha counting, with decay/ingrowth corrections), (also, gamma-spectrometry, 10 g).

^{228}Ra - (e) (1,5) (β-counting of ^{228}Ac).

^{210}Pb - (d) (1) (β-counting of ^{210}Bi).

^{210}Po - Ag deposition in presence of ascorbic acid and thioacetamide. (^{208}Po tracer used.)

^{232}Th, ^{228}Th, ^{230}Th - a series of iron hydroxide and barium sulphate coprecipitation steps followed by SX (TTA), back extraction (2 M HNO_3), coprecipitation with cerous hydroxide with collection on a 0.1 μm filter of α-spectrometry. (^{234}Th tracer). (^{232}Th also by NAA).

Lab-6 Decompositions: for Ra by fusion with Rushing flux, $Na_2CO_3/K_2CO_3/Na_2B_4O_7$ (10). For Th, Pb by KF and pyrosulphate fusions (11). For Po by multiple acid digestion; HF, HNO_3, H_2SO_4, HCl (1-2 g samples).

Separations:

^{226}Ra - ^{222}Rn emanated after 6 d. Storage of de-emanated solution in a sealed bubbler, ^{222}Rn and daughters measured by α-scintillation.

^{210}Pb - (d).

^{210}Po - Ni deposition from HCl medium (^{208}Po tracer, α-spectrometry).

^{230}Th - thorium in supernate from (c) coprecipitated with titanium hydroxide at pH 14 and filtered for alpha spectrometry.

Lab-7 Decompositions: for Ra, Th - repeated mixed acid digestion with HNO_3, HF, H_2SO_4, followed by pyrosulphate fusion and dissolution in HCl. For Pb - acid digestion with HNO_3, $HClO_4$; residue treated with HF, H_2SO_4; dissolved in HCl. (All 1 g samples.)

Separations:

^{226}Ra - (c) (1), α-spectrometry.

^{210}Pb - (d) (1), β-counting of ^{210}Bi, no recovery tracer.

^{230}Th - coprecipitation with lanthanum as hydroxide and then fluoride followed by SX (TTA), back extraction (HNO_3) (1), coprecipitation with titanium hydroxide and filtration for alpha spectrometry.

Lab-8 Decompositions of 2 g samples by repeated acid digestion (HF, H_2SO_4) followed by pyrosulphate fusion and dissolution in 10% HCl.

Separations:

^{226}Ra - (c).

^{230}Th, ^{232}Th, ^{228}Th - IX (cation, loaded pH 2-3, cations eluted with 12 M HCl, Th then eluted with 0.5 M oxalic acid) (13). Oxalate destroyed with HNO_3, SX (TTA), back-extracted into 2 M HNO_3 and Th isotopes electro-deposited (14) for α-spectrometry. (^{234}Th tracer used.)

^{210}Pb - IX eluent (above) passed through anion IX (to remove U) and Bi extracted by SX (DDTC), evaporated on planchet for β-counting 20 min and 1 d after extraction.

^{228}Ra - (e).

^{210}Po - Ag deposition, pH 0.3, 95°C; α-spectrometry, no tracer.

(a) Reports of contractors entitled 'Radiochemical determinations for tailings reference materials' and references cited should be consulted for details (Table 1, Table 13).

(b) Abbreviations: IX, ion-exchange chromatrography; SX, solvent extraction; TTA, theonyltrifluoroacetone; NAA, neutron activation analysis, HDEHP, bis(2-ethlyhexyl) phosphoric acid; DDTC, diethyldithiocarbenate; EDTA, ethlyenediaminetetraacetic acid; DTPA, diethylenetriaminepentaacetic acid; AAS, atomic absorption spectrophotometry.

(c) Radium (and other radioisotopes) are coprecipitated with lead sulphate; the precipitate is dissolved alkaline EDTA (or DTPA) and radium is selectively coprecipitated with barium sulphate which is collected on a membrane filter for α-spectrometry or gross alpha counting. Barium-133, added at the decomposition stage, is usually used as a recovery tracer, employing gamma-ray spectrometry [e.g. (1)].

(d) Lead and bismuth carriers are added prior to decomposition; the dissolved sample is in 2M HCl. Bismuth SX (0.1% DDTC/$CHCl_3$); extracts evaporated, decomposed with HNO_3, and bismuth hydroxides precipitated (pH 8). The precipitate is dissolved in HCl, and BiOCl (precipitated by dilution) is filtered for β-counting of ^{210}Bi after decay of other β-emitters. Correction is applied for decay of ^{210}Bi, and recovery is traced by AAS measurements of Bi carrier (1b).

(e) The Ba(Ra) SO_4 from (a) is redissolved in alkaline DTPA following a 3 d ingrowth of ^{228}Ac. Ba(Ra)SO_4 is removed by precipitation and ^{228}Ac is separated by SX (HDEHP/alkanes), back-extraction into 1 M HNO_3 and coprecipitation with lanthanum oxalate which is filtered for β-counting (1,5).

Table 13 - References cited for radiochemical procedures

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1(b) Smithson, G., Fahri, M. and Petrow, M. "Radiochemical determination of lead-210 in environmental samples and samples resulting from uranium mining-milling operations"; ibid, Appendix B.

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12. Donaldson, E.M. "Methods for the analysis of ores, rocks and related materials"; Monograph 881, second edition"; Ottawa, Supply and Services Canada; 1982.

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14. Puphal, K.W. and Olsen, D.R. "Electrodeposition of alpha-emitting nuclides from a mixed oxalate-chloride electrolyte"; Anal Chem 44:284-289; 1972.

Table 14 - Calibration reference materials and recovery tracers used for radiochemical measurements

	SRM (uncertainty, %)* [Tracer]						
	^{226}Ra	^{210}Pb	^{210}Po	^{230}Th	^{232}Th	^{228}Th	^{228}Ra
Lab 1	NBS 4964-B (0.5) [^{133}Ba]	-	-	-	st. monazite (1.2) -[thorium]-		-[cf. ^{226}Ra]-
Lab 2	-	AM RBZ (2.3) [Bi]	AM RBZ (2.3) [st. addn ^{210}Po]	IPL (6.) [^{234}Th]	-	-	-
Lab 3	NBS 4957 (1.8) [^{133}Ba]	AM RBZ (2.5) [Bi]	AM PMS (5) [^{208}Po int. std.]	AM RLZ (3.5) [^{228}Th int. std]	AM TYS-1 (<5) [cf. ^{230}Th]	AM RLS (3.5) [cf. ^{230}Th]	AM TYS-1 (5) [cf. ^{226}Ra]
Lab 4	NBS 4957 (1.8) [^{133}Ba]	-	AM PMS (5) [^{208}Po int. std.]	[^{228}Th int. std.]	-thorium nitrate- [cf. ^{230}Th]	(cf. ^{230}Th]	'aged' ThO_2 [no sepn.]
Lab 5	NBS 4953-C (1.8) [^{133}Ba]	AM RBZ (2.3) [Bi]	AM RBZ (2.3) [^{208}Po]	AM TYS-1 (<5) [^{234}Th]			(cf. ^{226}Ra]
Lab 6	AM RAY-31 (5) [n.i.]	AM RBZ (1.9) [Bi]	AM PDZ (3.4) [^{208}Po]	USEPA 1838-3 (5.7) [^{234}Th]	-	-	-
Lab 7	NBS 4959 (1.3) [^{133}Ba]	AM RBZ (2.3) [n.i.]	-	CCRMP BL-5 (?) [n.i.]	-	-	-
Lab 8	AM RAY-31 (5) [^{133}Ba]	NEN NES-200F (5) [n.i.]	[n.i.]	-NEN NES-316 (4.5)-	[^{234}Th]		NEN NES-200F (5) [cf. ^{226}Ra]

*Abbreviations: AM, Amersham Corp., IPL, Isotope Product Laboratories; NBS, National Bureau of Standards (U.S.); NEN, New England Nuclear Ltd.; USEPA, U.S. Environmental Protection Agency; n.i., not indicated.

Table 15 - Results of analysis of variance, ^{238}U-series isotopes*

		^{230}Th	^{226}Ra	^{210}Pb	^{210}Po
UTS-1	Ratio	7.02	0.89	9.89 [2.09]	34.9
	$F_{.95}$	2.57	2.44	2.60 2.81	2.68
	f_b, f_w	6,21	7,23	6,20 5,17	5,21
UTS-2	Ratio	26.3	1.77	7.37	20.7
	$F_{.95}$	2.55	2.35	2.41	2.64
	f_b, f_w	6,22	7,29	6,31	5,23
UTS-3	Ratio	7.08	1.00	13.6 [1.19]	18.55
	$F_{.95}$	2.55	2.46	2.53 2.90	2.64
	f_b, f_w	6,22	7,22	6,23 5,15	5,23
UTS-4	Ratio	41.6 [5.58]	3.90	33.6	45.3
	$F_{.95}$	2.57 2.77	2.40	2.56	2.66
	f_b, f_w	6,21 5,18	7,25	6,21	5,22
UTS-5	Ratio	24.7	14.0	19.1	21.5
	$F_{.95}$	2.51	2.42	2.57	2.68
	f_b, f_w	6,24	7,24	6,21	5,21

*Ratio of 'between-set' to 'within-set' mean-square variance with degrees of freedom f_b and f_w, resp. (4).

[], value after removal of probable outlier set.

Table 16 - Results of analysis of variance, ^{232}Th-series isotopes*

		^{232}Th	^{228}Ra	^{228}Th
UTS-1	Ratio	2.92 [1.94]	7.32	2.34
	$F_{.95}$	2.61 3.18	3.71	3.05
	f_b, f_w	7,17 4,13	3,10	4,15
UTS-2	Ratio	3.14 [2.88]	33.9	17.8
	$F_{.95}$	2.58 3.11	3.59	3.01
	f_b, f_w	7,18 4,14	3,11	4,16
UTS-5	Ratio	6.12 [7.87]	13.52	16.4
	$F_{.95}$	2.61 3.18	3.06	3.05
	f_b, f_w	7,17 4,13	4,15	4,15

[] alpha spectrometry methodology only.

Table 17 - Results of Dixon r tests applied to laboratory mean results

Material	Isotope	Set	No sets	Test ratio	Critical Value, r	
					P = 10%	P = 5%
UTS-1	^{210}Pb	Lab-7	7	.61	.51	.57
UTS-1	^{228}Ra	Lab-5*	4	.90	.77	.82
UTS-3	^{210}Pb	Lab-5	7	.52	.51	.57
UTS-4	^{230}Th	Lab-3	7	.71	.51	.57

*Test result overruled; set means of the other three sets were judged to be fortuitously close.

Table 18 - Consensus values and related statistical parameters for isotope activities in tailings reference materials

Isotope	Consensus value, [CL], Bq g^{-1} [a.] [No. sets, No. values, RSD (%), $\overline{CV}$(%)]			
	UTS-1	UTS-2	UTS-3	UTS-4
^{230}Th	3.6	4.4	11.3	22.9[b]
	[3.0 - 4.2]	[3.3 - 5.5]	[10.5 - 12.1]	[20.3 - 25.5]
	(7,28,21,10)	(7,29,26,24)	(7,29,9,5)	(6,24,13,7)
^{226}Ra	3.67	5.6	13.3	38.6
	[3.52 - 3.82]	[5.2 - 6.0]	[12.7 - 13.9]	[36.2 - 40.9]
	(8,31,9,7)	(8,37,13,8)	(8,30,11,6)	(8,33,9,5)
^{210}Pb	3.36[b]	4.6	12.6[b]	34.7
	[3.15 - 3.58]	[3.9 - 5.3]	[11.7 - 15.5]	[30.4 - 39.0]
	(6,23,9,7)	(7,38,16,10)	(7,30,14,7)	(7,28,12,4)
^{210}Po	3.1	4.4	11.8	30.8
	[2.7 - 3.5]	[3.7 - 5.1]	[10.8 - 12.9]	[25.8 - 35.9]
	(6,27,10,4)	(6,29,14,6)	(6,29,8,4)	(6,28,14,4)
^{232}Th[c,d]	.68	.88	(.16 ± .04)	(.48 ± .06)
	[.59 - .77]	[.67 - 1.08]		
	(5,18,16,11)	(5,19,24,17)		
^{228}Ra	.68	1.0	-	-
	[.47 - .89]	[0.7 - 1.4]		
	(4,14,19,10)	(4,15,17,6)		

Table 18 (Cont'd)

Isotope	Consensus value, [CL], Bqg^{-1} [a.] [No. sets, No. values, RSD (%), $\overline{CV}$(%)]			
	UTS-1	UTS-2	UTS-3	UTS-4
^{228}Th [d]	.71	.92	(.16 ± .04)	(.23 ± .04)
	[.58 - .84]	[.58 - 1.25]		
	(5,20,21,13)	(5,21,29,11)		
^{231}Pa [d]	(.21 ± .03)	(.37 ± .05)	(.70 ± 1.0)	(2.4 ± .3)

a. CL = statistical uncertainty range at 95% confidence level, RSD = relative standard deviation of individual result (%), $\overline{CV}$ = average within-laboratory rel. std. dev. (%).

b. Data are exclusive of an outlying set.

c. ^{232}Th results from alpha spectrometry only. ^{232}Th concs. by NAA are incorporated with chemical results.

d. Single laboratory results and uncertaintly estimates in brackets are for information only.

Table 19 - Radioisotope consensus program performance on UTS-5
[DH-1a/SiO_2, 49.4 ± .6% ore]

Isotope	Consensus, Bq g^{-1} Mean	95% CL	CI, ± %*	Bias Estimate %**	No. Sets	No. values	RSD %	$\overline{CV}$, %
^{230}Th	15.4	14.1 - 16.7	8.5	-2.9	7	31	9.1	3.5
^{226}Ra	16.4	14.9 - 17.8	8.8	+3.2	8	32	10.9	3.9
^{210}Pb	15.0	12.7 - 17.3	15.2	-5.4	7	28	15	6.0
^{210}Po	13.6	11.4 - 15.9	16.6	-14.2	6	27	15	7.0
^{232}Th(a)	1.94	1.72 - 2.15	11.2	+6.7	8	25	14	6.4
^{232}Th(b)	1.96	1.59 - 2.34	19.2	+7.8	5	18	16	7.9
^{228}Ra	1.80	1.14 - 2.47	37.3	-1.1	5	20	27	15.3
^{228}Th	1.98	1.60 - 2.37	19.5	+8.9	5	20	16	6.7
^{231}Pa	(.58)	(.55 - .61)	(5)	-22.	1	4	-	12.9

*one-sided 95% confidence interval, %.

**100 $(x_c - x_p)/x_p$ where x_p and x_c are predicted and consensus values, resp. Uncertainties in the bias estimates due to concentration and half-life uncertainties of uranium and thorium resp. are approx. ±2.1 and ±5.3.

a) all data.

b) α-spectrometry results only.